

REMARKS

Reconsideration of this application is respectfully requested. Claims 1, 2 and 7 have been withdrawn from consideration. Claim 6 has been cancelled without prejudice or disclaimer. Claims 3 and 5 have been amended. Claims 8-11 have been newly added. No new matter has been added by this Amendment. Upon entry of this Amendment, claims 1-5, 7 and 8-10 are pending and claims 3-5 and 8-11 are currently at issue.

Claim amendments

Claim 3 has been amended to more particularly describe the invention, to recite each step in the gerund form, to explicitly recite an alkaline earth metal, and to call for the addition of a zeolite seed crystal. Support for these amendments can be found in the specification, for example, on page 7, lines 7-13, page 8, lines 3-9 and page 10, lines 15-23, and original claim 6. No new matter has been added by these amendments. In addition, claim 3 has been amended to call for an "aqueous gel mixture" formed in dissolving step (I). Support for this amendment can be found on page 8, lines 9-11 of the specification. No new matter has been added by this amendment.

Claim 5 has been amended to remove multiple dependencies and to recite each step in the gerund form. No new matter has been added by this amendment. New claim 8 recites the subject matter of claim 5 without multiple dependency. Support for this amendment can be found in the claims as originally filed. No new matter has been added by this amendment.

New claims 9 and 10 call for specific organic and inorganic alkaline earth metal salts, respectively. Support for these claims can be found throughout the specification, for example on page 6, lines 17-120 and Example 1, page 10, line 15. No new matter has been added by these amendments.

New claim 11 calls for a specific zeolite seed crystal. Support for this claim can be found in Example 1, page 10, lines 20-21 of the specification. No new matter has been added by this amendment.

Rejections Under 35 U.S.C. § 112

Claims 3-5 stand rejected under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention. The Examiner states that the terminology "an alkaline earth metal salt and a zeolite seed crystal are coexistent with each other" is indefinite. This ground for rejection is not well taken. It is respectfully submitted that amended claim 1 does not call for the term "an alkaline earth metal salt and a zeolite seed crystal are coexistent with each other."

The Examiner also states that there is no antecedent basis for the zeolite seed crystal "which is added" in claim 3. It is respectfully submitted that amended claim 3 calls for the addition of a zeolite seed crystal in dissolving step (I), thereby providing the proper antecedent basis for the zeolite seed crystal "which is added" in claim 3.

The claim terminology in claims 3-5 clearly meet the strictures of the second paragraph of 35 U.S.C. § 112 and the Examiner is requested to withdraw the rejection of these claims.

Rejections Under 35 U.S.C. § 103

Claims 3-5 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Japanese Patent No. JP 60-251121 ("the '121 patent") in view of Japanese Patent No. JP 61-058812 ("the '812 patent"). The Examiner states that the ZSM-5 zeolite catalyst of the '121 patent is produced from a mixture analogous to the mixture called for by the present claims, and that the ranges of the catalyst ingredients in the '121 patent are commensurate with those of the present claims. The Examiner also contends that it is well known in the art of zeolite synthesis to employ seed crystals in order to produce final compositions having crystals of consistent and desired particle size. The Examiner further contends that the '812 patent discloses ZSM-5 seed crystals "on the order of those claimed" in the present application, and therefore it would have been obvious to employ the '812 crystals in the process taught by the '121 patent.

First, it is respectfully submitted that the ZSM-5 zeolite catalyst of the '121 patent is not produced from a mixture analogous to the mixture called for by the present claims. A partial English translation of the '121 patent is attached as Exhibit A. The present invention calls for an alkaline earth metal to be present when the mixture from step (I) is subjected to hydrothermal treatment in step (II) of claim 3. The process disclosed by the '121 patent, on the other hand, does not disclose an alkaline earth metal in the mixture subjected to hydrothermal treatment (See claim 1, claim 2 and the section labeled page 4, right-upper column, line 8 to right-lower column, line 1 of the partial English translation of the '121 patent attached as Exhibit A).

Second, it is respectfully submitted that even if it were obvious to use of the '812 seed crystal in the process taught by the '121 patent, this would not achieve the process called for by claim 3 of the present invention because the '121 process does not comprise an alkaline earth metal in the hydrothermal treatment step. A partial English translation of the '812 patent is attached as Exhibit B. It is respectfully submitted that the use of the seed crystal of the '812 patent in the '121 process would not result the process called for by claim 3 of the present invention because the resulting process would lack the presence of an alkaline earth metal in the mixture during the hydrothermal treatment in step (II) as called for by the present claims.


Therefore, the combination of the seed crystal of the '812 patent with the process of the '121 patent does not make up for the deficiency of the '121 process, namely the lack of an alkaline earth metal in the mixture subjected to hydrothermal treatment.

For the foregoing reasons claim 3 of the present invention is not obvious in view of the combined teachings of the '121 patent and the '812 patent. Claims 4 and 5 depend from claim 3 and are therefore also not obvious over the combination of the '121 patent and the '812 patent. The Examiner is therefore respectfully requested to withdraw the rejection of claims 3-5 for obviousness over the '121 patent in view of the '812 patent.

In view of the preceding comments and amendments, the pending claims are believed to be in condition for allowance and such action is earnestly solicited.

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Exhibit A - Partial Translation of the '121 Patent

S 60-251121

Page 1, left-column, line 4 to line 16:

--2. Claims:

(1) A method for preparing a microcrystalline ZSM-5 type zeolite, wherein when an aqueous solution containing a silica source, an alumina source, an alkali source, and a quaternary alkyl ammonium salt is heated under reflux at a normal pressure, the molar ratio $\text{SiO}_2/\text{H}_2\text{O}$ of the silica source and water is defined in the range from 5 to 20, and the heating under reflux is continued for 6 to 13 days.

(2) A method for preparing a microcrystalline ZSM-5 type zeolite (including silicalite), wherein when a solution containing a silica source, an alkali source, and a quaternary alkyl ammonium salt is heated under reflux at a normal pressure, the molar ratio $\text{SiO}_2/\text{H}_2\text{O}$ of the silica source and water is defined in the range from 5 to 20, and the heating under reflux is continued for 6 to 13 days.--

Page 4, right-upper column, line 8 to right-lower column, line 1:

--Example 1

In this Example, silica sol Cataloid SI-30 (SiO_2 : 30 wt%, H_2O : 70 wt%) that is commercially available from Catalysts & Chemicals Industries Co., Ltd. was chosen as an SiO_2 source, a commercially available special grade reagent $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ was chosen as an Al_2O_3 source, a commercially available special grade reagent NaOH was chosen as an alkali source, and a commercially available special grade reagent tetra-n-propylammonium bromide (TPA) was chosen as an organic crystallizing agent. A gel solution of the starting raw material mixture was prepared in the following method.

158.4g of Cataloid SI-30 was collected into a polypropylene Erlenmeyer flask equipped with a Teflon-made magnetic stirrer, having an internal volume of 500 ml, and 1.698 g of $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, 10.2 g of NaOH, and 10.8 g of TPA were added in this order to the solution under stirring. The pH of the white turbid homogeneous gel solution is about 13.5 at room temperature, and the molar ratios of each composition of the starting mixture are the injection ratios as follows:

$$\text{SiO}_2/\text{Al}_2\text{O}_3 = 350$$

$$\text{OH}/\text{SiO}_2 = 0.322$$

$$\text{TPA}/\text{SiO}_2 = 0.0513$$

$$\text{H}_2\text{O}/\text{SiO}_2 = 7.83.$$

Next, the Erlenmeyer flask into which the starting mixture had been put was equipped with a reflux cooler, and the contents of the Erlenmeyer flask were heated under reflux and stirring for 11 days in an oily bath (set at 110°C) equipped with a magnetic stirrer. The obtained product was repeatedly washed with water and separated in the mother liquid by a centrifuge, and the size of the crystal particles was measured by phase identification by X-ray diffraction measurement (XRD) using a CuK α -ray and Scanning Electron Microscopy (SEM).

As a result of XRD, the obtained product showed a typical diffraction pattern of Na-TPA-ZSM-5 type zeolite. In addition, it could be seen that the average crystal particle diameter as determined by SEM was about 0.3 μm , and a submicron-order scale ZSM-5 type zeolite microcrystalline was obtained by this method.--

Exhibit B - Partial Translation of the '821 patent

S 61-58812

Page 1, left-column, line 5 to page 2, left-upper column, line 8:

--2. Claims

(1) A method for preparing a ZSM-5 type crystalline aluminosilicate zeolite, comprising crystallizing a raw material mixture containing a silica, an alumina, an alkali metal, an organic cation, and water by heating, wherein upon crystallization, a crystalline aluminosilicate powder having the X-ray diffraction pattern shown in Table below, obtained by crystallizing a raw material mixture containing no organic cation, is allowed to be present in the raw material mixture.

(2) The method as described in claim 1, wherein the crystalline aluminosilicate powder having the X-ray diffraction pattern shown in Table is present at 0.01 to 5% by weight of the raw material mixture.

(3) The method as described in claim 1, wherein the crystalline aluminosilicate powder having the X-ray diffraction pattern shown in Table has an $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 20 to 30.--

Page 3, right-upper column, line 4 to right-lower column, line 17:

-- The crystalline zeolite powder contained in the raw material mixture as the seed crystal of the present invention is obtained by reacting organic-cation free raw material mixture, and is essentially crystalline aluminosilicate powder having the X-ray diffraction pattern shown in Table 1. This seed crystal has $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio ranging from 15 to 35. However, those which are most likely to have the above X-ray diffraction pattern are those having the

molar ratio ranging from 20 to 30. The additive amount of the seed crystal preferably ranges from 0.01 to 5 weight % to the raw material mixture.

This seed crystal can be prepared, for example, in the manner to be described below. A raw material mixture, which includes white carbon as a silica source, sodium aluminate as an alumina source, and sodium hydroxide and water as alkali conditioner, is prepared. Additionally, in a further method, a raw material mixture, in which an amorphous aluminosilicate homogeneous phase compound obtained by reacting an aqueous sodium silicate solution and an aqueous aluminum-containing solution simultaneously or continuously is dispersed in an aqueous sodium hydroxide solution, is prepared. It is a feature of the present invention that the raw material mixture has no organic cation. This raw material mixture has a composition as shown in terms of each oxide molar ratio:

$$\text{SiO}_2/\text{Al}_2\text{O}_3 = 20 \text{ to } 45$$

$$\text{Na}_2\text{O}/\text{SiO}_2 = 0.05 \text{ to } 0.3$$

$$\text{H}_2\text{O}/\text{SiO}_2 = 12 \text{ to } 70.$$

After stirring the raw material mixture at room temperature until it turns sufficiently homogeneous, it is kept at a temperature of 120°C to 200°C for 5 hours to 5 days under stirring in an autoclave to carry out crystallization. Next, it is subjected to solid-liquid separation, sufficiently washed, and dried at 110°C overnight.

The crystalline zeolite thus obtained is a crystalline zeolite having an $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 20 to 30 and the X-ray diffraction pattern shown in Table 1, which would become a seed crystal usable in the present invention.

It is one of the features of the X-ray diffraction pattern of the seed crystal that although synthesis is conducted without the use of an organic mineralizing agent, only two peaks are observed between the lattice plane spacing $d=3.70 \text{ \AA}$ and $d=3.86 \text{ \AA}$. For ZSM-5 which is synthesized with the addition of organic mineralizing agent, 2 to 3 peaks are observed between the lattice plane spacing $d=3.70 \text{ \AA}$ and $d=3.86 \text{ \AA}$, as disclosed in Japanese

Patent Publication No. 46-10064. However, for the X-ray diffraction pattern of the ZSM-5-like zeolite which is synthesized with no addition of an organic mineralizing agent and with the use of silica sol as a silica source, as disclosed in Japanese Patent Publication No. 56-49851, 4 peaks are observed between the lattice plane spacing $d=3.70 \text{ \AA}$ and $d=3.86 \text{ \AA}$. That is, the seed crystal used in the present invention is a crystalline aluminosilicate zeolite which is completely different in nature from the ZSM-5-like zeolite as disclosed in Japanese Patent Publication No. 56-49851.

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Page 4, right-upper column, line 18 to left-lower column, line 7:

-- If ZSM-5 is synthesized using the seed crystal according to the present invention having the characteristics as above, ZSM-5 having a particle diameter of 0.1 to 1.0 micron is obtained. Additionally, surprisingly, the nucleation of ZSM-5 occurs vigorously on the particle surface of the seed crystal, the nucleus grows only to a size of at most 0.5 micron. In addition, it could be seen that the seed crystal did not grow at all. Moreover, it was also surprised that the seed crystal and the particle of ZSM-5 are separated by a later washing step, which thus leads to homogeneous ZSM-5 having a particle diameter of 1 micron or less.--

Page 4, left-lower column, line 16 to right-lower column, line 5:

-- Therefore, the seed crystal of the present invention has property which is quite different from ordinary ZSM-5 or ZSM-5 like zeolite. That is, the seed crystal which can be used in the present invention is that which is obtained by crystallizing raw material mixture of organic cation-free, having the X-ray diffraction pattern shown in Table 1, i.e. only crystalline aluminosilicate powder having an $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 15 to 35. The seed crystal is allowed to be present at 0.01 to 5% by weight of the raw material mixture.--